

Light scattering by wavelength-sized particles “dusted” with subwavelength-sized grains

Michael I. Mishchenko,^{1,*} Janna M. Dlugach,² and Daniel W. Mackowski³

¹NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA

²Main Astronomical Observatory of the National Academy of Sciences of Ukraine, 27 Zabolotny Street, 03680, Kyiv, Ukraine

³Department of Mechanical Engineering, Auburn University, Auburn, Alabama 36849, USA

*Corresponding author: mmishchenko@giss.nasa.gov

Received November 17, 2010; revised December 10, 2010; accepted December 13, 2010;

posted December 14, 2010 (Doc. ID 138269); published January 25, 2011

The numerically exact superposition *T*-matrix method is used to compute the scattering cross sections and the Stokes scattering matrix for polydisperse spherical particles covered with a large number of much smaller grains. We show that the optical effect of the presence of microscopic dust on the surfaces of wavelength-sized, weakly absorbing particles is much less significant than that of a major overall asphericity of the particle shape. © 2011 Optical Society of America

OCIS codes: 290.0290, 290.1090, 290.5825, 290.5850.

Comparisons of laboratory measurements of light scattering by nonspherical particles and theoretical computations often show significant disagreements. The origin of these disagreements can ultimately be traced to the perennial inability to model electromagnetic scattering by exact replicas of actual particle ensembles, partly caused by the lack of an appropriate numerical tool to provide precise results using reasonable computational power and within realistic computation time. It is, therefore, very important to analyze, both qualitatively and quantitatively, what specific morphological features of particles can have distinct and significant effects on light scattering.

It is well recognized that many natural and artificial particles exhibit both overall nonsphericity of shape and small-scale surface irregularities. The latter can manifest themselves in the form of microscopic pits and knobs as well as microscopic grains “dusting” the surfaces of the larger host particles [1–3]. While the effects of overall nonsphericity of wavelength-sized particles on the optical cross sections and Stokes scattering matrix have been documented extensively and have been demonstrated to be quite significant (e.g., [4–6] and references therein), the effects of microscopic surface irregularities are much less understood, the main reason being the inadequacy of the existing theoretical tools. There have been attempts to analyze the effects of surface imperfections on light-scattering characteristics of particles with sizes substantially greater than the incident wavelength [7–11]. However, the underlying theoretical technique (geometric optics) is approximate by definition and cannot be applied reliably to wavelength-sized particles. Several recent studies have been based on numerically exact computer solvers of the Maxwell equations [12–19], but they were largely limited to monodisperse rather than polydisperse particle models and did not consider the potential optical effects of small grains contaminating the surfaces of much larger host particles. Furthermore, in many cases the roughening algorithm used obviously affected not only the particle surface but also the overall particle shape.

In this Letter we use the numerically exact and efficient superposition *T*-matrix method [20,21] in order

to analyze the likely optical effects of microscopic grains dusting the surfaces of wavelength-sized particles. Since we are not interested in the effects of a major asphericity of the host particles, we use a simple and computationally affordable scattering model in the form of a large sphere covered with 49 much smaller grains (Fig. 1). The critical advantage of this model is that it allows one to separate unequivocally the scattering effects of the host shape and those of the host being dusted with small grains.

The ratio of the size parameter of the host to that of the small grains is kept constant at $X/x \equiv 10$. The small grains are placed on the surface of the large host using a random-number generator, making sure that the grains do not overlap. The scattering characteristics are then averaged over the uniform orientation distribution of the resulting host-grain's configuration as well as over a standard power-law distribution $n(X) = \text{constant} \times X^{-3}$ [5] with an effective size parameter $X_{\text{eff}} = 10$, effective variance $v_{\text{eff}} = 0.05$, and host size parameters in the range $X \in [6.61, 14.39]$. The refractive index of the large host and small surface grains is fixed at $1.55 + i0.0003$, which is a value typical of mineral particles. The numerical averaging over size is based on a Gaussian integration formula with 100 quadrature points. Sample computations for a few host size parameters have shown that different arrangements of the 49 small grains on the surface of the large host yield essentially the same values of the resulting optical observables as long as these arrangements are sufficiently random. Therefore, the same (appropriately scaled) spatial configuration is used for all 100 size parameters entering the numerical integration.

Figure 1 visualizes the elements of the normalized Stokes scattering matrix relating the Stokes parameters of the incident quasi-monochromatic light and those of the scattered light in the far-field zone:

$$\begin{bmatrix} I^{\text{sca}} \\ Q^{\text{sca}} \\ U^{\text{sca}} \\ V^{\text{sca}} \end{bmatrix} \propto \begin{bmatrix} a_1(\Theta) & b_1(\Theta) & 0 & 0 \\ b_1(\Theta) & a_2(\Theta) & 0 & 0 \\ 0 & 0 & a_3(\Theta) & b_2(\Theta) \\ 0 & 0 & -b_2(\Theta) & a_4(\Theta) \end{bmatrix} \begin{bmatrix} I^{\text{inc}} \\ Q^{\text{inc}} \\ U^{\text{inc}} \\ V^{\text{inc}} \end{bmatrix}, \quad (1)$$

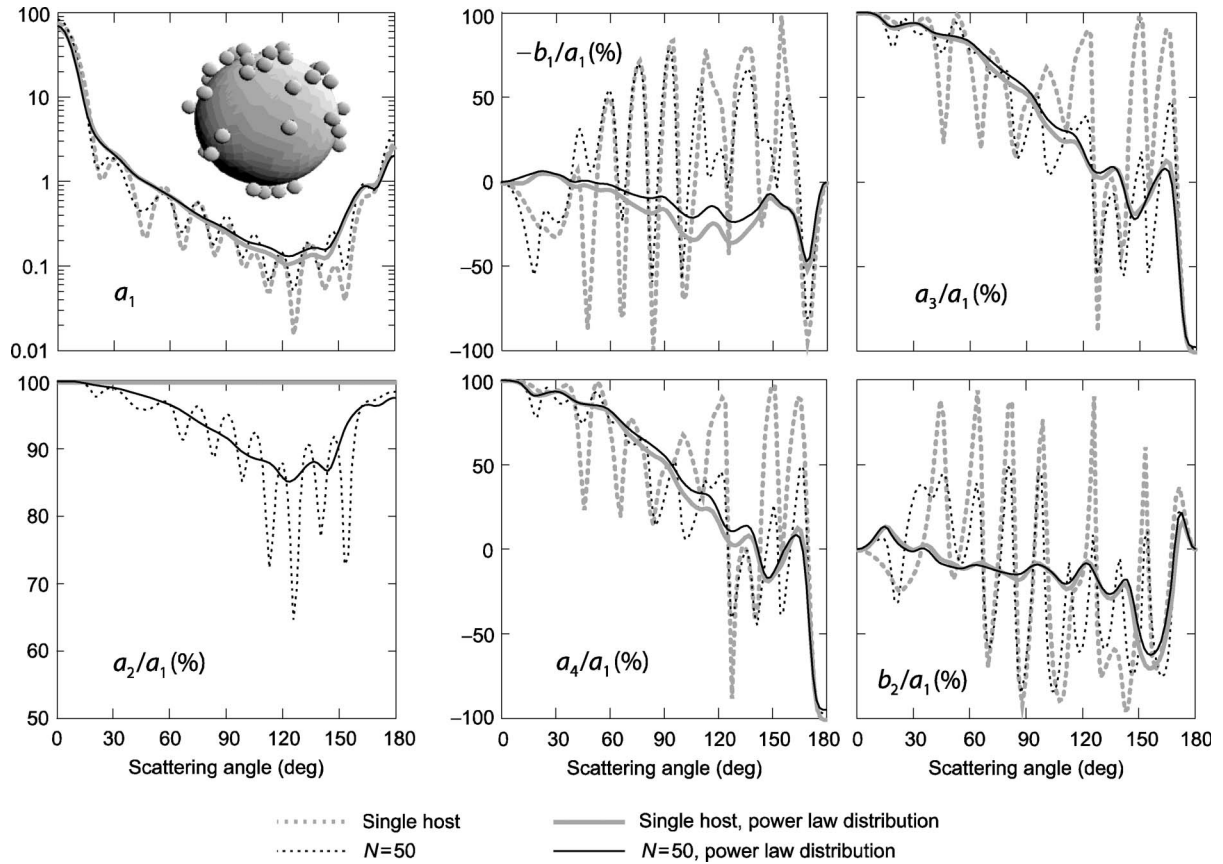


Fig. 1. Elements of the normalized Stokes scattering matrix for a single monodisperse host, a monodisperse host dusted with 49 small particles, a polydisperse host, and a polydisperse host dusted with 49 small particles. The inset shows the host–dust grains configuration used in our T -matrix computations.

where $\Theta \in [0^\circ, 180^\circ]$ is the angle between the incidence and scattering directions, and both sets of the Stokes parameters are defined with respect to the scattering plane [5]. The (1,1) element is the conventional phase function normalized according to

$$\frac{1}{2} \int_0^\pi d\Theta \sin \Theta a_1(\Theta) = 1. \quad (2)$$

In theory, the off-block diagonal elements of the scattering matrix would be zero only if the multisphere configurations studied had a plane of symmetry [22]. We found, however, that in the cases considered the maximum magnitude of the minor matrix elements was much smaller than that of the major elements. The results in Fig. 1 are shown for monodisperse ($X = 10$, $x = 1$) as well as polydisperse ($X_{\text{eff}} = 10$, $x_{\text{eff}} = 1$, $v_{\text{eff}} = 0.05$) cases. For comparison, the results for “uncontaminated” monodisperse and polydisperse hosts are also displayed.

It should be emphasized that these data are the result of a direct, numerically exact computer solution of the macroscopic Maxwell equations and involve no approximations beyond an appropriate truncation of the infinite T matrix to a finite, numerically converged size. As such, they demonstrate, first of all, that it may be highly problematic to decipher the effects of particle microphysics on scattering based on monodisperse results alone (cf. [5,23]). Indeed, the black and gray dashed curves in Fig. 1 can exhibit significant differences, whereas the so-

lid polydisperse curves reveal a much closer agreement. Second, the polydisperse results in Fig. 1 suggest that the optical effects of microscopic irregularities on the surfaces of wavelength-sized hosts are rather weak, if not negligible. This conclusion is corroborated by the close agreement of the corresponding polydisperse single-scattering albedos (0.9926 and 0.9923 for the contaminated and uncontaminated hosts, respectively) and asymmetry parameters (0.661 and 0.668). The effect of the small dust grains on the forward-scattering diffraction peak is also vanishingly small.

It is, thus, obvious that the optical effects of surface contamination are much weaker than those of a major overall departure of the particle shape from that of a perfect sphere. Perhaps the only unequivocal indicator of nonsphericity of the scattering object shown in Fig. 1 is the noticeable deviation of the ratio $a_2(\Theta)/a_1(\Theta)$ from 100% [5]. Among the less pronounced effects are the weak enhancement of the phase function at side-scattering angles and the equally weak suppression at backscattering angles coupled with somewhat more neutral linear polarization $-b_1(\Theta)/a_1(\Theta)$ at side-scattering angles. Although these effects are qualitatively similar to those observed, e.g., for randomly oriented polydisperse spheroids and cylinders in comparison with volume- or surface-equivalent spheres (e.g., [4–6] and references therein), their small magnitude makes them much less significant.

We expect that our results will be important in analyses of laboratory and remote-sensing data intended to identify the likely causes of differences in light-scattering properties of particles with various morphologies. At least in the case of nonabsorbing or weakly absorbing wavelength-sized particles, our numerically exact computations are indicative of a rather weak optical effect of microscopic surface irregularities in comparison with the effects of a major overall asphericity. For practical reasons, the power-law size distribution used in our computations is rather narrow, and yet it is capable of averaging off most of the monodisperse differences. Therefore, averaging over a wider (and, potentially, more realistic) size distribution can be expected to yield even smaller polydisperse differences between “clean” and “dusty” hosts. Admittedly, it would also be interesting to analyze the case when the surface of a large host particle is covered with small grains entirely or almost entirely. Unfortunately, the limited efficiency of the computer facilities currently available to us makes this task problematic.

There is no reason to expect that our main conclusion should change in the case of host particles with much larger size parameters as long as the typical size-parameter scale of irregularities remains of order unity. However, this conclusion should not be extrapolated to strongly absorbing particles (cf. [19]) and/or large particles with much larger surface irregularities. Unfortunately, the efficiency of the existing numerically exact solvers of the Maxwell equations in the geometric-optics domain of size parameters remains limited, which makes problematic a reliable quantitative analysis of the effects of wavelength-sized and larger surface imperfections on light scattering.

We are grateful to Timo Nousiainen for a useful discussion and anonymous reviewers for constructive comments and suggestions. This research was supported by the National Aeronautics and Space Administration (NASA) Radiation Sciences Program managed by Hal Maring and by the NASA Glory Mission Project.

References

1. H. Volten, O. Muñoz, E. Rol, J. de Haan, W. Vassen, J. Hovenier, K. Muinonen, and T. Nousiainen, *J. Geophys. Res.* **106**, 17375 (2001).
2. O. Muñoz and H. Volten, in *Light Scattering Reviews: Single and Multiple Light Scattering*, A. A. Kokhanovsky, ed. (Springer-Praxis, 2006), pp. 3–29.
3. T. Nousiainen, *J. Quant. Spectrosc. Radiat. Transfer* **110**, 1261 (2009).
4. M. I. Mishchenko, L. D. Travis, R. A. Kahn, and R. A. West, *J. Geophys. Res.* **102**, 16831 (1997).
5. M. I. Mishchenko, L. D. Travis, and A. A. Lacis, *Scattering, Absorption, and Emission of Light by Small Particles* (Cambridge University, 2002), <http://www.giss.nasa.gov/~crim/books.html>.
6. O. Dubovik, A. Sinyuk, T. Lapyonok, B. N. Holben, M. Mishchenko, P. Yang, T. F. Eck, H. Volten, O. Muñoz, B. Veihelmann, W. J. van der Zande, J.-F. Leon, M. Sorokin, and I. Slutsker, *J. Geophys. Res.* **111**, D11208 (2006).
7. A. Macke, J. Mueller, and E. Raschke, *J. Atmos. Sci.* **53**, 2813 (1996).
8. P. Yang and K. N. Liou, *Contrib. Atmos. Phys.* **71**, 223 (1998).
9. P. Yang, Q. Feng, G. Hong, G. W. Kattawar, W. J. Wiscombe, M. I. Mishchenko, O. Dubovik, I. Laszlo, and I. N. Sokolik, *J. Aerosol Sci.* **38**, 995 (2007).
10. K. Muinonen, T. Nousiainen, H. Lindqvist, O. Muñoz, and G. Videen, *J. Quant. Spectrosc. Radiat. Transfer* **110**, 1628 (2009).
11. B. A. Baum, P. Yang, Y.-X. Hu, and Q. Feng, *J. Quant. Spectrosc. Radiat. Transfer* **111**, 2534 (2010).
12. M. I. Mishchenko and A. A. Lacis, *Appl. Opt.* **42**, 5551 (2003).
13. W. Sun, T. Nousiainen, K. Muinonen, Q. Fu, N. G. Loeb, and G. Videen, *J. Quant. Spectrosc. Radiat. Transfer* **79–80**, 1083 (2003).
14. C. Li, G. W. Kattawar, and P. Yang, *J. Quant. Spectrosc. Radiat. Transfer* **89**, 123 (2004).
15. T. Rother, K. Schmidt, J. Wauer, V. Shcherbakov, and J.-F. Gayet, *Appl. Opt.* **45**, 6030 (2006).
16. T. Nousiainen and K. Muinonen, *J. Quant. Spectrosc. Radiat. Transfer* **106**, 389 (2007).
17. E. Zubko, K. Muinonen, Y. Shkuratov, G. Videen, and T. Nousiainen, *J. Quant. Spectrosc. Radiat. Transfer* **106**, 604 (2007).
18. J.-C. Auger, G. E. Fernandes, K. B. Aptowicz, Y.-L. Pan, and R. K. Chang, *Appl. Phys. B* **99**, 229 (2010).
19. M. Kahnert, T. Nousiainen, and P. Mauno, “On the impact of small-scale surface roughness and non-sphericity on the optical properties of hematite aerosols,” *J. Quant. Spectrosc. Radiat. Transfer* (to be published).
20. D. W. Mackowski and M. I. Mishchenko, *J. Opt. Soc. Am. A* **13**, 2266 (1996).
21. D. Mackowski, K. Fuller, and M. Mishchenko, <ftp://ftp.eng.auburn.edu/pub/dmckowski/scatcodes/index.html>.
22. H. C. van de Hulst, *Light Scattering by Small Particles* (Wiley, 1957).
23. J. E. Hansen and L. D. Travis, *Space Sci. Rev.* **16**, 527 (1974).